Application No. 10/574, 907

## PROPOSED AMENDMENT TO CLAIM 1

1. (Currently Amended) A film-type catalyst for production of a tertiary amine, which is used in producing a tertiary amine from an alcohol and a primary or secondary amine as the starting material;

wherein said film-type catalyst comprises catalyst particles bound to one another via a binder;

wherein said particles form a three-dimensional network structure via the binder;

wherein said film-type catalyst has a thickness of 500  $\mu m$  or less and a pore volume of not less than 0.5 mL/m<sup>2</sup>,

whereby the diffusion rate in the catalyst layer is increased <u>due to said three-dimensional</u> <u>network structure</u>, and the mass transfer between the inside and outside of the catalyst can be promoted thereby utilizing the whole of the catalyst and simultaneously suppressing the excessive reaction of [[the]] <u>an</u> intermediate reaction product in the inside of the catalyst.

Support for the above amendment to claim 1 occurs in the original specification, such as paragraphs [0030] to [0038] of US 2008/0004472 A1 (reproduced below).

[0030] The film-type catalyst can contain a binder which does not act as an active substance but fixes the active substance to form the film-type catalyst. The binder includes high-molecular or inorganic compounds which are durable to the reaction environment, do not adversely affect the reaction system and have not only a property of binding particles of the active substance together or binding the active substance to the surface of a substance but also properties such as chemical resistance and heat resistance. Example of the binder include cellulose resin such as carboxymethyl cellulose and hydroxyethyl cellulose, fluorine-based resin such as poly(tetrafluoroethylene) and poly(vinylidene fluoride), high-molecular compounds such as urethane resin, epoxy resin, polyester resin, phenol resin, melamine resin, silicone resin, polyvinyl alcohol, polyimide resin and polyimide amide resin, and sol of inorganic compound such as silica and alumina.

[0031] The internal structure of the film-type catalyst depends significantly on the type of the active substance constituting the catalyst and on the method if preparing the catalyst, and may form a dense continuous phase or may be porous. For example, when the catalyst is a thin film formed in the surface of a substrate by sputtering and chemical vapor deposition, the catalyst can have a dense continuous phase, while when the powdery active substance is used to form a thin on the surface of a substrate by wet or dry coating, the catalyst can be porous.

[0032] As the method of producing the film-type catalyst, it is particularly preferable to use a method of forming, on a substrate, a coating containing a powdery catalyst active substrate and a binder for fixing it. Particularly, in the film-type catalyst obtained by forming, on a substrate, a coating film containing a powdery catalyst together with synthetic resin used as a binder in the method of producing the same, the compounding ratio of the powdery catalyst to the synthetic resin can be regulated in such a suitable range as to control the degree of exposure of the powdery catalyst and the binding strength among the catalyst powders and the binding strength between the substrate and the catalyst powders.

[0033] In the film making method, the compounding ratio is preferably a ratio of 20 to 80 parts by weight of the synthetic resin to 100 parts by weight of the powdery catalyst. The film-type catalyst exhibiting the reaction activity particularly effective as a film-type catalyst and hardly releasing its coating can be obtained by further controlling the pore volume per unit area of the substrate in the range of 0.5 to 30 mL/m<sup>2</sup>.

[0034] In the internal structure of the film-type catalyst obtained by the above film making method, the surface of the powdery catalyst is coated wholly or partially with the synthetic resin, and the powdery catalyst particles are bound to one another via the synthetic resin, and a large number of the powdery catalyst particles are deposited in the direction of thickness. A large number of such deposited powdery catalyst particles form a 3-dimensional network structure via the synthetic resin, and air layers lie scattered in spaces among the powdery catalyst particles, and preferably the synthetic resin itself also has a pore structure. The film-type catalyst can have a gradient structure wherein the ratio of the powdery catalyst to the synthetic resin is changed in the direction of thickness, or the film-type catalyst layer may include two or more catalyst layers different in the ratio of the powdery catalyst to the synthetic resin therein two or more catalyst layers different in pore structure.

[0035] In the film-type catalyst obtained by the film making method described above, the synthetic resin is contained preferably in an amount of 20 to 80 parts by weight relative to 100 parts by weight of the powdery catalyst, whereby the catalyst activity, or the binding strength among the powdery catalyst particles or between the powdery catalyst and the substrate, can be effectively exhibited. In the film-type catalyst, the pore volume of the catalyst layer per unit surface area of the substrate is desirably 0.5 to 30 mL/m², preferably 1 to 20 mL/m², more preferably 1 to 4 mL/m². When the pore volume is not less than 0.5 mL/m², the diffusion rate in the catalyst layer is increased, and upon application of the film-type catalyst to the reaction process, a high reaction rate can be obtained. When the pore volume is too large, the catalyst layer is made porous at high degree, the binding strength among the powdery catalyst particles and /or between the powdery catalyst and the substrate is reduced thus reducing the mechanical strength of the catalyst layer, while when the pore volume is 30 mL/m² or less, the porosity of

the catalyst layer becomes suitable, and the adhesion among the powdery catalyst particles and/or between the powdery catalyst and the substrate is increased. As a result, the mechanical strength of the catalyst layer is increased and simultaneously the mass transfer of the reactants and products can be kept in an excellent state.

[0036] The pore structure of the film-type catalyst obtained by the film making method is determined in a step of eliminating volatile components and in step of forming a twined structure of synthetic resin after coating a substrate with a coating containing the synthetic resin preferably in the range of 20 to 80 parts by weight relative to 100 parts by weight of the powdery catalyst. When the synthetic resin is thermosetting resin, the pore structure is determined at the time of forming a crosslinked structure (network structure) formed by progress of curing/crosslinking reaction, in addition to the step of eliminating volatile components. When the reaction is accompanied by condensation reaction, the pore structure is also influenced by the step of eliminating the condensation product. As a result of eager study, it was found that when the pore volume per unit surface area if the substrate, as an indicator characterizing the pore structure formed, is 0.5 to 30 mL/m², sufficient reaction rate of the film-type catalyst in the reaction process and the retaining properties of the powdery catalyst can be preferably maintained.

[0037] The powdery catalyst used in the film making method is preferably a catalyst whose performance has been established for the suspension-bed process. In this case, the development time for film-type catalyst, as compared with the method of converting a catalyst active substance precursor fixed on a substrate into a catalyst active substance by treatment such as calcination, can be shortened because reaction characteristics in the reaction process can be secured. The production process can be simplified because the complicated production process for exhibiting the reaction activity, such as calcination treatment, is not necessary.

[0038] The powdery catalyst used in film making method is preferably the one having particle diameters, preferably with sharp distribution, of 0.01 to 500  $\mu$ m, preferably 0.1 to 100  $\mu$ m, more preferably 0.5 to 50  $\mu$ m, in terms of average particles diameter, and a specific surface area of 0.1 to 5000 m²/g, preferably 1 to 200 m²/g, more preferably 10 to 100 m²/g, as determined by the BET method. For example, the powdery catalyst can be prepared by using the following catalyst carrier and catalyst active substance precursor.